

## Photoinduced Emission of Cooper Pairs from Superconductors

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Under certain conditions specified in this work, a Cooper pair can be emitted from a superconductor upon the absorption of one ultraviolet photon. The spectra of the excited electron pair carry direct information on the energy and angular pair correlation. These statements are concluded from a formal and numerical analysis based on the Bardeen-Cooper-Schrieffer theory for superconductivity.

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The properties of conventional weak-coupling superconductors (SCs) are well described by the theory put forth by Bardeen, Cooper, and Schrieffer (BCS) [1]: Below a critical temperature  $T_c$  the electron-phonon interaction correlates two electrons close to the Fermi level  $E_F$  to a Cooper pair (CP) [2], a bound two-electron state with vanishing total wave vector and spin. The result of this pairing is a slight lowering of the total energy of the system and the opening of a narrow gap of the order of a few meV in the electronic density of states around  $E_F$ , inhibiting thus the kind of collisions which lead to ordinary resistivity; hence, for temperatures  $T$  such that the thermal energy is less than the band gap, the material shows no resistivity [3]. In contrast to this simple scenario for (conventional) superconductivity, the mechanisms underlying high-temperature superconductivity (HTSC) [4] are complex and not yet completely understood (cf., e.g., [5] and references therein). However, there are indications for the relevance of the pairing concept to the emergence of superconductivity at elevated temperatures [6].

The properties of superconductive materials, such as the magnitude, the symmetry, and the temperature dependence of the superconducting gap, have been studied by various experimental techniques. Here we mention the neutron and the polarization-resolved Raman spectroscopy [7,8], the infrared photoabsorption spectroscopy [9], the de Haas–van Alphen effect [10], and the scanning tunneling spectroscopy [11–13]. A technique of particular importance for exploring the properties of SCs is the photoemission spectroscopy (PES) [14,15] where, upon the excitation of the material by a monochromatic radiation, electrons are emitted and analyzed with respect to their wave vector distribution. In the superconducting state, an energy gap  $2\Delta(T)$  (centered around  $E_F$ ) occurs in the angular integrated energy spectrum of the photoelectron [see inset of Fig. 2(b) (below)]. Within a pairing theory, this gap is related to the minimal energy required to break the CP resulting in two unpaired electrons in the supercondensate [3]. Since only one of the two electrons is detected, PES does not allow one to study the angular and/or energy pair correlation. For an insight in how the electrons are correlated, one has to utilize a method

capable of resolving simultaneously the quantum states of *two* electrons following the excitation of a CP.

In this Letter, we suggest the use of the ultraviolet one-photon two-electron coincidence spectroscopy [also called double-photoelectron (DPE) spectroscopy] for the study of the properties of superconductors. Thanks to spectacular recent advances in coincidence techniques, the DPE spectroscopy has been utilized for studying correlation effects in nuclear [16], atomic [17], and molecular [18] systems. Recently, the DPE from condensed matter has been realized [19]. In DPE, the correlation within the pair is mapped out by fixing the wave vector of one of the detected electrons while scanning the wave vector of the other. A distinctive feature is that DPE is prohibited if the electrons are spatially uncorrelated [20], which makes this technique predestinate for studying correlation induced phenomena, such as superconductivity. In view of the advances in experimental techniques, it is timely to develop the first theory for DPE from SCs and to calculate and analyze the DPE spectra. This Letter provides a general theory for DPE from superconductors using the two-particle Green's function approach [21]. The theory is capable of treating PES and DPE allowing thus a comparison of the two techniques. Pilot numerical calculations are performed for DPE from conventional, weak-coupling superconducting samples that are well described by the BCS mean field theory. We singled out this case (and not a HTSC) to avoid an ambiguity in the interpretation of the DPE spectra that may originate from the employed mechanism of superconductivity, even though, as argued below, DPE is potentially useful to unveil some of the secrets of the HTSC phase.

*General formulation.*—We consider a process (Fig. 1) where, upon the absorption of a photon with energy  $\omega$ , two electrons are emitted from a SC sample into the vacuum where their energies  $E_{1/2}$  and emission angles  $\theta_{1/2}$  are determined (energies are measured relative to the chemical potential, and atomic units are used). The state  $p_j \equiv \mathbf{p}_j \sigma_j$  of the electron  $j$  is specified by the electron wave vector ( $\mathbf{p}_j$ ) and spin ( $\sigma_j$ ). The DPE current  $J_{12}$  is expressible in terms of the particle-particle Green functions and the hole-hole spectral function  $A_{12}^{(-)}$  as [21,22]

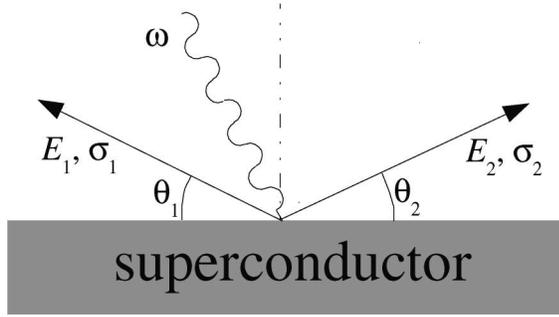


FIG. 1. Schematics of the DPE process indicating the photon energy ( $\omega$ ), and the energies ( $E_1, E_2$ ) and spins ( $\sigma_1, \sigma_2$ ) as well as the emission angles ( $\theta_1, \theta_2$ ) of the two electrons.

$$J_{12} \propto \langle \Psi_{p_1 p_2}^{(-)} | D_{12} A_{12}^{(-)}(E_{12}) D_{12}^\dagger | \Psi_{p_1 p_2}^{(-)} \rangle \\ = \sum_{k_1 k_2 k'_1 k'_2} M_{k_1, k_2} M_{k'_1, k'_2}^\dagger A_{12}^{(-)}(k_1 k_2 k'_1 k'_2; E_{12}). \quad (1)$$

The matrix elements  $M_{k_1, k_2}$ , given by

$$M_{k_1, k_2} = \langle \Psi_{p_1 p_2}^{(-)} | D_{12} | k_1 k_2 \rangle, \quad (2)$$

are formulated in a spinor plane-wave basis  $|k_1 k_2\rangle \equiv |k_1\rangle \otimes |k_2\rangle$ . In Eq. (1),  $E_{12} = E_1 + E_2 - \omega$  is the binding energy of the occupied two-electron state, and  $D_{12} = (A/c)[e^{i\mathbf{q}\cdot\mathbf{r}_1} \hat{\mathbf{e}} \cdot \mathbf{p}_1 + e^{i\mathbf{q}\cdot\mathbf{r}_2} \hat{\mathbf{e}} \cdot \mathbf{p}_2]$  is the coupling operator of the photon to the electron pair. Here we assumed a monochromatic, plane-wave vector potential ( $\mathbf{A}$ ) with a polarization vector  $\hat{\mathbf{e}}$  and a wave number  $\mathbf{q}$ .  $c$  is the light velocity and  $\mathbf{p}_{1/2}$  are the one-electron momentum operators. The correlated photoelectron-pair state  $|\Psi_{p_1 p_2}^{(-)}\rangle$  is obtained by back propagating (in the presence of the crystal potential) the asymptotic (detected) pair state  $|p_1 p_2\rangle$  by means of the particle-particle propagator.

*Pair states in a superconductor.*—A determining factor

$$A_{\text{UP}}^{(-)}(E_{12}) = (\delta_{k_1, k'_1} \delta_{k_2, k'_2} - \delta_{k_1, k'_2} \delta_{k_2, k'_1}) [(1 - n_{k_1})(1 - n_{k_2}) v_{k_1}^2 v_{k_2}^2 \delta(E_{12} + E_{k_1} + E_{k_2}) + n_{k_1} n_{k_2} u_{k_1}^2 u_{k_2}^2 \delta(E_{12} - E_{k_1} - E_{k_2}) \\ + (1 - n_{k_1}) n_{k_2} v_{k_1}^2 u_{k_2}^2 \delta(E_{12} + E_{k_1} - E_{k_2}) \\ + n_{k_1} (1 - n_{k_2}) u_{k_1}^2 v_{k_2}^2 \delta(E_{12} - E_{k_1} + E_{k_2})] \quad (6)$$

describes two independent Bogobolons. For  $T > T_c$ ,  $A_{\text{UP}}^{(-)}$  reduces to

$$A_{\text{UP}}^{(-)}(E_{12}) = (\delta_{k_1, k'_1} \delta_{k_2, k'_2} - \delta_{k_1, k'_2} \delta_{k_2, k'_1}) f_{k_1} f_{k_2} \\ \times \delta(E_{12} - \varepsilon_{k_1} - \varepsilon_{k_2}),$$

which describes two independent normal state elementary excitations [ $f_k = 1/[\exp(\beta\varepsilon_k) + 1]$ ]. Hence, we associate  $A_{\text{UP}}^{(-)}$  with uncorrelated electron-pair (UP) states. The part of the spectral function

$$A_{\text{CP}}(E_{12}) = \delta_{k_1, -k_2} \delta_{k'_1, -k'_2} \frac{\Delta_{k_1}(T) \Delta_{k'_1}(T)}{4E_{k_1} E_{k'_1}} \delta(E_{12}) \quad (7)$$

corresponds to the condensed CPs [ $\Delta_k(T)/2E_k$  is the wave function of CP in momentum space] and vanishes above

of the CP photocurrent (1) is the hole-hole spectral function  $A_{12}^{(-)}$ . Considering a conventional SC sample at temperatures  $T < T_c$ , we employ the BCS mean field theory and evaluate  $A_{12}^{(-)}$  (in momentum space) as

$$A_{12}^{(-)}(k_1 k_2 k'_1 k'_2; E_{12}) = \int \frac{dt}{2\pi} e^{-iE_{12}t} \langle \langle c_{k'_1}^\dagger(t) c_{k'_2}^\dagger(t) c_{k_2} c_{k_1} \rangle \rangle. \quad (3)$$

Here  $c_k^\dagger(t)$  [ $c_k(t)$ ] is the creation [annihilation] operator (in the Heisenberg representation) of an electron with quantum numbers  $k \equiv \mathbf{k}\mu$  ( $\mathbf{k}$  is the wave vector and  $\mu$  is the spin). The thermodynamical average  $\langle \langle \dots \rangle \rangle$  is calculated using the Bogoliubov operators  $\alpha_k^+(t) = e^{iE_k t} \alpha_k^+$  [ $\alpha_k(t) = e^{-iE_k t} \alpha_k$ ] that create [annihilate] elementary excitation (Bogobolon) with energy  $E_k = \sqrt{\varepsilon_k^2 + \Delta_k^2(T)}$ , where  $\varepsilon_k$  is the excitation energy in the normal state and  $\Delta_k(T)$  is the energy gap in the SC state [note that in DPE we are dealing with a high-energy pair excitation of SC which is fundamentally different from the known (particle-hole) Bogoliubov spectrum]. The Bogoliubov operators are related to the electron operators through the unitary transformation (see, for example, Ref. [3])

$$c_k^+ = u_k \alpha_k^+ + v_k \alpha_{-k}, \quad (4)$$

where the expansion (coherence) factors satisfy the relations

$$v_k^2 + u_k^2 = 1, \quad v_k = \frac{1}{2} \left[ 1 - \frac{\varepsilon_k}{E_k} \right], \quad u_k v_k = \frac{\Delta_k(T)}{2E_k}.$$

After some algebra and integrating over the time variable, we deduce from (3) and (4) that

$$A_{12}^{(-)}(k_1 k_2 k'_1 k'_2; E_{12}) = A_{\text{UP}}^{(-)}(E_{12}) + A_{\text{CP}}(E_{12}), \quad (5)$$

where [we define  $n_k = 1/[\exp(\beta E_k) + 1]$ ,  $\beta = 1/k_B T$ ]

$T_c$ . The dependence of  $A_{\text{CP}}$  on the pairing mechanism is encompassed in the gap function relation  $\Delta_k = \frac{1}{2} \sum_{k'} V(k, k') (\Delta_{k'}/E_{k'})$  [where  $V(k, k')$  is the pairing potential]. The splitting of  $A_{12}^{(-)}$  [Eq. (5)] in an uncorrelated (6) and a correlated part (7) is useful since the probability for the (incoherent) emission of two uncorrelated electrons (e.g., by two photons) is governed by a function of the form (6). This incoherent emission constitutes a part of the background for the coincidence experiment and, hence, it is important to inspect its structure.

*DPE current from SC state.*—Given the structure of Eq. (5), the DPE current consists of two components:

$$J_{12} = J_{\text{UP}} + J_{\text{CP}}, \quad (8)$$

where

$$J_{\text{UP}} \propto \sum_{k_1 k_2} (|M_{k_1, k_2}|^2 - M_{k_1, k_2} M_{k_2, k_1}^*) [(1 - n_{k_1})(1 - n_{k_2}) v_{k_1}^2 v_{k_2}^2 \delta(E_{12} + E_{k_1} + E_{k_2}) + n_{k_1} n_{k_2} u_{k_1}^2 u_{k_2}^2 \delta(E_{12} - E_{k_1} - E_{k_2}) + (1 - n_{k_1}) n_{k_2} v_{k_1}^2 u_{k_2}^2 \delta(E_{12} + E_{k_1} - E_{k_2}) + n_{k_1} (1 - n_{k_2}) u_{k_1}^2 v_{k_2}^2 \delta(E_{12} - E_{k_1} + E_{k_2})], \quad (9)$$

$$J_{\text{CP}} \propto \delta(E_{12}) \sum_{kk'} M_{k, -k} M_{k', -k'}^* \frac{\Delta_k(T) \Delta_{k'}(T)}{4E_k E_{k'}}. \quad (10)$$

The matrix elements for a given pair state  $(k_1, k_2)$  depend on the photon polarization  $\hat{\mathbf{e}}$ , the photon wave vector  $\mathbf{q}$ , as well as on the photoelectron-pair state  $(p_1, p_2)$ , and encompass the spin and wave vector selection rules for the possible electron-pair transitions  $(k_1, k_2) \rightarrow (p_1, p_2)$ . The matrix element (2) has the following general properties. (i) It is nonzero only if the states  $(k_1, k_2)$  and  $(p_1, p_2)$  are in the same spin states  $(\mu_{1(2)} = \sigma_{1(2)})$ , for the transition operator  $D_{12}$  is spin independent. (ii) For crystalline samples, the condition  $(\mathbf{p}_1 + \mathbf{p}_2)_{\parallel} + \mathbf{g}_{\parallel} = (\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{q})_{\parallel}$  applies, where  $\mathbf{g}_{\parallel}$  is the surface reciprocal lattice vector. Mathematically, the conditions (i) and (ii) can be expressed as

$$M_{k_1, k_2} = m_{\mathbf{k}_1, \mathbf{k}_2} \delta[(\mathbf{p}_1 + \mathbf{p}_2)_{\parallel} + \mathbf{g}_{\parallel} - (\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{q})_{\parallel}] \delta_{\sigma_1 \mu_1} \delta_{\sigma_2 \mu_2}, \quad (11)$$

where  $m_{\mathbf{k}_1, \mathbf{k}_2}$  is a reduced matrix element. (iii) In metallic SCs, the (micron-size) coherence length  $\xi$  (the extent of the CP) is much larger than the screening length  $\lambda$  (few lattice constants). Thus, unlike the atomic and molecular case, the Coulomb electrostatic repulsion of the electrons is subsidiary for DPE from SCs. As a consequence, if the *initial pair state* has an *s*-wave character, the photoelectron-pair current (8) and the matrix element do not depend dynamically on the polarization of the photon [23]. This fact can be utilized to map out the non-isotropic components of the initial state by measuring the normalized circular dichroism  $\text{CD} = [J_{12}(\text{LH}) - J_{12}(\text{RH})]/[J_{12}(\text{LH}) + J_{12}(\text{RH})]$ , where  $J_{12}(\text{LH}/\text{RH})$  is the photoelectron-pair current induced by the absorption of a left-hand (right-hand) circularly polarized photon. In fact, a direct relation between CD and the phase of the CP state can be derived. Thus, the measurement of CD is most relevant for HTSC materials to unravel the gap symmetry properties. The numerical examples shown below are restricted to *s*-wave pairing where CD plays no role.

*Correlated energy spectrum.*—The CP has vanishing wave vector and spin, therefore for its emission the relations  $\sigma_1 = -\sigma_2$  and  $(\mathbf{p}_1 + \mathbf{p}_2)_{\parallel} + \mathbf{g}_{\parallel} = \mathbf{q}_{\parallel}$  should apply. As deduced from Fig. 1, the photoelectron pair may be emitted in the plane perpendicular to the surface with  $E_1 = E_2 = E$ ,  $\sigma_1 = -\sigma_2 = \sigma$ , and  $\theta_1 = \theta_2 = \theta$ . In this case only the states  $(k_1, k_2)$  satisfying  $\mu_1 = -\mu_2$ ,  $(\mathbf{k}_1 + \mathbf{k}_2)_{\parallel} = 0$  can contribute to the DPE current. The energy spectrum of the photoelectron-pair can be studied as a

function of  $E$  in two regimes: (i)  $\theta = \text{const}$  or (ii)  $\theta \neq \text{const}$ ,  $|\mathbf{p}_{1,\parallel}| = |\mathbf{p}_{2,\parallel}| = \text{const}$ . In both regimes, the matrix elements vary only weakly with  $E$  in the range of several  $\Delta'$  (note  $\Delta < 5$  meV and  $\xi \gg \lambda$ ). As shown in Fig. 1, we do not investigate here the azimuthal dependence  $\varphi_{1/2}$  on the electron-pair emission directions since we will be considering isotropic *s*-wave pairing. For materials (such as HTSC) with nonisotropic gap, the dependence on  $\varphi_{1/2}$  is of a prime interest.

To demonstrate the characteristic features of  $J_{\text{CP}}$ , we use Eq. (11) and assume  $m_{\mathbf{k}_1, \mathbf{k}_2}$  to be constant on the energy scale of few  $\Delta'$ . For numerical illustrations, we replace the energy and momenta Dirac functions in Eqs. (9)–(11) by Lorentzians with half widths at half maximum  $0.1\Delta$  and  $0.1k_F$ , respectively. A free-electron model is assumed for the normal state, as done in the single photoelectron spectroscopy [15]. The six-dimensional integration over momenta is performed analytically in Eq. (10) and fully numerically in Eq. (9). Figure 2 shows the results for correlated DPE spectrum in the case of *s*-wave pairing [ $\Delta_k(0) = \Delta_0 \theta(\omega_D - |\varepsilon_k|)$ , where  $\omega_D$  is the Debye frequency]. As depicted in Fig. 2(a), a pronounced peak occurs at zero binding energy  $E_{12}$  in the DPE current from CP states. The peak position and strength reflect, respectively, the condensation and the (macroscopic) number of CPs in the system. Above  $T_c$ , only the uncorrelated DPE current  $J_{\text{UP}}$  is present. As seen in Fig. 2(b), this current is modified

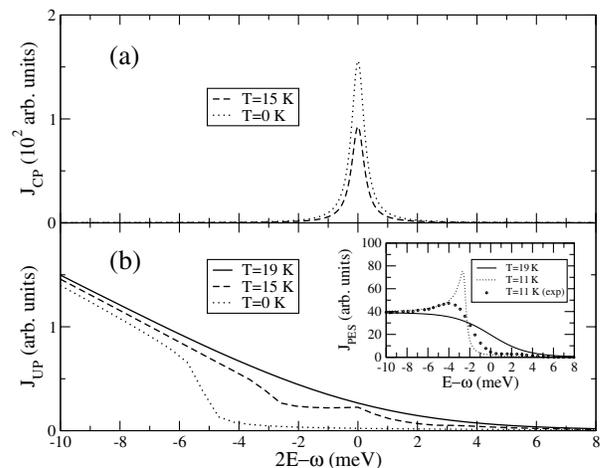


FIG. 2. DPE current from  $\text{V}_3\text{Si}$  ( $\Delta_0 = 2.6$  meV,  $T_c = 17$  K) as a function of the energy  $2E - \omega = E_{12}$  and temperature. Panels (a) and (b) show  $J_{\text{CP}}$  [Eq. (10)] and  $J_{\text{UP}}$  [Eq. (9)]. Theoretical results and experimental data [15] for the angle-integrated single photoemission are included in the (b) inset.

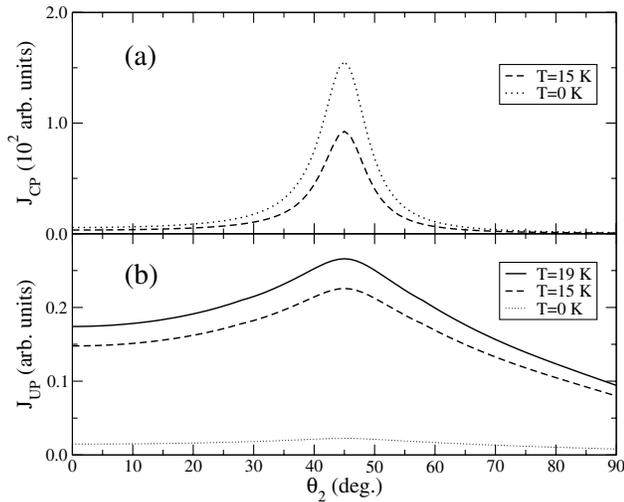


FIG. 3. The electron-pair angular distributions from  $V_3Si$  as a function of  $\theta_2$  at fixed  $\theta_1 = 45^\circ$ ,  $E_1 = E_2 = \omega/2 \approx 10.63$  eV (see Fig. 1). In (a) the contribution  $J_{CP}$  [Eq. (10)] is shown, whereas in (b)  $J_{UP}$  [Eq. (9)] is displayed.

below  $T_c$  due to the opening of a gap in the one-electron density of states (that can be measured in PES). The theoretical PES results in Fig. 2(b) are obtained using the BCS model (see Ref. [15] for details) and the same energy broadening as in the DPE case. A comparison of these results with Fig. 2(a) shows that below  $T_c$  the DPE energy spectrum exhibits a qualitatively different feature: a peak at  $E_{12} = 0$  instead of a shift towards  $E_{12} < 0$  as in PES. The reason is that in PES the absorption of a photon creates an unpaired hole in the supercondensate, whereas in DPE it creates two paired holes in the supercondensate.

*Correlated angular distributions.*—The angular distributions of  $J_{CP}$  and  $J_{UP}$  are shown, respectively, in Figs. 3(a) and 3(b) ( $E_1 = E_2 = \omega/2$ , i.e.,  $E_{12} = 0$ ). All distributions exhibit a maximum at  $\theta_2 = \theta_1 = 45^\circ$ , when  $(\mathbf{p}_1 + \mathbf{p}_2)_\parallel = 0$ . However, for the CP states this peak is sharper and is orders of magnitude larger than for  $J_{UP}$ , which is a consequence of the condensation of CPs in the pair state with zero sum momentum. Basically, the physical parameters governing the magnitude of  $J_{CP}$  are readily inferred from Eq. (10). However, as the results shown in Figs. 2 and 3 are obtained upon a six-dimensional integration with a finite life time of the states, an analytical estimate of  $J_{CP}/J_{UP}$  is not possible. Our observation is that a smaller lifetime increases the ratio  $J_{CP}/J_{UP}$ .

*Possible experimental realization.*—The present analysis is relevant to the case of conventional weak-coupling superconductors that are well described within the BCS model. The according experimental studies may be hampered by the small value of the energy gap in these materials (usually  $\sim 1$  meV) as compared to the best achievable energy resolution ( $\sim 1$ – $3$  meV [24]). A suitable BCS material is the A15-type conventional SC  $V_3Si$  with a gap of  $\Delta \approx 2.5$  meV. Here detailed high-

resolution PES studies have been conducted [15] [cf. Figure 2]. HTSC materials are obviously more accessible experimentally (energy gap is typically  $\sim 10$  meV and higher [25]). In addition, for HTSC materials the coherence length  $\xi$  (the CP size) can be comparable to the screening length  $\lambda$ , which is favorable for enhancing the magnitude of the DPE matrix elements [see Eq. (11)].

DPE from HTSCs has the potential for providing a novel insight into pair correlations, pseudogap effects, the gap energy, and symmetry (using circularly polarized photons), as well as into the temperature dependence of these quantities.

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